Disorder Induced Cluster Formation near First Order Phase Transitions in Electronic Systems: Importance of Long-Range Coulomb Interaction

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We discuss the effects of fluctuations of the local density of charged dopants near a first order phase transition in electronic systems, that is driven by change of charge carrier density controlled by doping level. Using a generalization of the Imry-Ma argument, we find that the first order transition is rounded by disorder at or below the lower critical dimension $d_c = 3$, when at least one of the two phases has no screening ability. The increase of d_c from 2 (as in the random field Ising model) to 3 is due to the long-range nature of the Coulomb interaction. This result suggests that large clusters of both phases will appear near such transitions due to disorder, in both two and three dimensions. Possible implications of our results on manganites and underdoped cuprates will be discussed.

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Study of effects of quenched disorder on first order phase transitions has a long history. The prototype model for such studies is the random field Ising model (RFIM) [1]. At low-temperature the pure Ising model spontaneously develops magnetization and there are two different phases corresponding to the two possible directions of the magnetization. A *uniform* external field h drives the systems through a first order phase transition at $h_c = 0$, where the magnetization changes discontinuously. If a *random* field with zero mean is applied on the other hand, it would like the local order parameter to point in the direction of the local field, thus clusters or domains of both phases would form, the global magnetization would be destroyed, and the first order transition associated with the jump of global magnetization would be rounded. In order for this to happen however, the energy gain due to forming clusters with magnetization following the local random field direction, E_h, must be large enough to overcome the energy penalty associated with the domain walls separating the clusters with opposite magnetization, E_w. Imry and Ma [1] analyzed the competition between these two effects as a function of the linear size of the cluster L, and found that for systems with dimensionality d less than the lower critical dimension $d_c = 2$, $E_h \sim L^{d/2}$ overwhelms $E_w \sim L^{d-1}$ for large enough L; in this case clusters will form at sufficiently large length scales *no matter how weak the random field is*, and the first order transition is rounded. For $d > d_c$ on the other hand the global magnetization and first order transition remain intact for *weak* random field. For $d = d_c = 2$, E_h and E_w scale with L the same way: $E_h \sim E_w \sim L$. More detailed studies [2,3] indicated that at $d = d_c = 2$, E_h eventually dominates E_w at large L and destroys the ordered phase and the associated first order transition. These considerations and conclusions have been generalized to generic first order phase transitions [4].

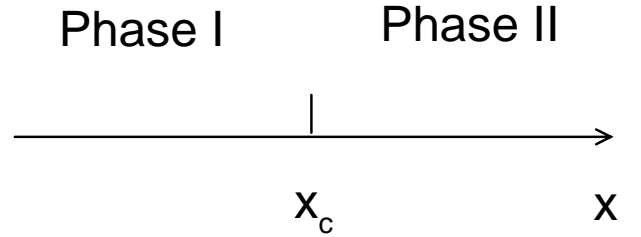Recently there has been considerable interest in the

FIG. 1. Schematic phase diagram of an electronic system. As the doping level x is varied, there are two phases separated by a first order phase transition at $x = x_c$.

rounding of first order phase transitions in electronic systems, and the associated cluster formation. In these systems the first order transition in the absence of disorder is driven by the change of charge carrier density, which in turn is controlled by the density of dopants introduced into the system; see Fig. 1. Here the dopants play a dual role. First of all they provide the charge carriers (conduction electrons or holes), and their density determines the location of the system in the phase diagram. On the other hand they are also the source of disorder, as they distort the local lattice structure and *more importantly, there is local fluctuation in their density*. As we argue below, due to the long-range nature of the Coulomb interaction between the dopants and the charge carriers, the effect of dopant density fluctuation is much stronger than the random field in the random field Ising model; the latter only couples to the *local* order parameter and thus represents short-range interaction. A consequence of that is it increases the lower critical dimension d_c from 2 to 3; we thus expect in both 2- and 3-dimensional systems the otherwise first-order phase transitions are rounded by dopant density fluctuations, and clusters of both phases will always form near the phase boundary. In the following we illustrate these points by performing an extension of the original Imry-Ma analysis. We will con-

sider two different generalizations of the Coulomb interactions in 3D to arbitrary dimensions. The first is to keep the momentum dependence: $V(q) \sim 1/q^2$, which yields $V^{(1)}(r) \sim r^{2-d}$ in real space (in 2D, $V^{(1)}(r) \sim \log r$); the other is simply using $V^{(2)}(r) \sim 1/r$ in all dimensions. We will discuss both cases.

Now assume that the *average* dopant density of the system is $\overline{n} = n_c$, *i.e.* at the transition point (or doping level $x = x_c$ in Fig. 1). In the *absence* of any disorder, the system will be in one of the two phases depicted in Fig. 1 and there are no clusters or domains, since generically there is an energy penalty associated with the domain walls that is proportional to the area of the walls. We now discuss what happens when there is local fluctuation of the dopant density. Consider a domain of linear size L. The average number of dopants in this domain is $N(L) \sim L^d$, while the typical *fluctuation* of $N(L)$ is $\delta N(L) \sim \sqrt{N(L)} \sim L^{d/2}$. Obviously, the local fluctuation of dopant density tend to force domains of both phases to form, according to the local density of dopants; the energy penalty of *not* forming domains is dominated by the electrostatic energy due to the imbalance between the dopant charge and carrier charge in the regions that are in the "wrong" phase where the carrier density cannot reach that of the local dopant density, which is

$$E_c(L) \sim [\delta N(L)]^2 V(L) \sim L^d V(L) \tag{1}$$

per domain. For the two types of generalization of Coulomb interaction, we find

$$E_c^{(1)}(L) \sim L^2 \tag{2}$$

and

$$E_c^{(2)}(L) \sim L^{d-1} \tag{3}$$

respectively. In order for the domain formation to occur, the gain of Coulomb energy E_c must overcome the domain wall energy penalty

$$E_w \sim L^{d-1}. \tag{4}$$

Comparing the expression of E_w with those of $E_c^{(1)}$ and $E_c^{(2)}$, we find for the first generalization of Coulomb interaction, $E_c^{(1)}$ dominates E_w at sufficiently large L for $d < d_c = 3$, *i.e.*, the lower critical dimension above which the first order transition remains intact is $d_c = 3$; at 3D the effect of the dopant density fluctuation is marginal. For the second generalization of Coulomb interaction, $E_c^{(2)}$ scales with L exactly the same way as E_w, *regardless of the dimensionality d.* Thus the effects of dopant density fluctuation is always marginal.

These results are very different from that of the random field Ising model (RFIM), in which case $d_c = 2$; the origin of the fundamental difference lies in the long-range nature of the Coulomb interaction, which yields very different dependence of E_c on the domain size L, as compared with $E_h \sim L^{d/2}$ of the RFIM.

The discussions presented above are quite general. In the following we propose a simple lattice model that contains the basic ingredients of the physics being considered here, which can be studied numerically to check the results obtained above [5]:

$$H = \frac{1}{2} \sum_{ij} \frac{(\rho_i - n_i)(\rho_j - n_j)}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{J}{2} \sum_{<ij>} [1 - sgn(\rho_i \cdot \rho_j)]. \tag{5}$$

Here i and j are indices of lattice sites of a d-dimensional cubic lattice, n_i is the quenched background (or dopant) charge which follows some specific distribution like Gaussian or box distributions, and ρ_i is the electron charge which is the dynamical variable of the model, and it is varied to minimize H. The first term represents the $1/r$ Coulomb interaction among the charge carriers, and the summation is over all pairs of sites in the system; it can also be replaced by other forms of long-range interaction that is appropriate. The second term represents domain wall energy cost and the sum is over nearest neighbors; here without losing generality we have assumed the phase boundary is located at $\rho_c = 0$, thus there is an energy cost of J for each nearest neighbor bond that connects sites with opposite signs of ρ, or sites occupied by different phases (the function sgn returns the sign of its argument). It is clear that in the absence of fluctuation in the background (or dopant) charge density, *i.e.*, $n_i = n$ independent of i, we will simply have $\rho_i = n$ everywhere, there is no competition between the two terms in H, and there is a first order transition at $n = 0$ from the phase with $\rho < 0$ to the phase with $\rho > 0$. On the other hand when n_i follows certain distribution with a finite width, and its average $\overline{n}$ is close to 0, these two terms compete with each other. The first term prefers to have ρ_i match n_i everywhere, thus ρ_i with both signs (or both phases) would appear in the system; the fluctuation of n_i plays a role similar to the random field in the RFIM. On the other hand the second term penalizes near neighbor pairs with opposite signs of ρ; J plays a role similar to the Ising coupling in the RFIM. Numerical simulation of this and other closely related models are currently underway [6].

A few comments on our results are now in order.

(i) It is very important in our discussion that the first order transition is driven by the change of doping level. There are, of course, other first order transitions in electronic systems that are driven by other mechanisms (like change of pressure) *without* changing the doping level; our results do not apply to those cases, as long as the position of the first order transition does not involve the doping level.

(ii) Within the RFIM, it is known that if there exists power-law correlation in the random field, the lower

critical dimension d_c may be increased [7]. The increase of d_c due to long-range Coulomb interaction discussed here has some resemblance to this effect, but the precise mechanism of increase of d_c is different.

(iii) In the case of the RFIM, it has been proven rigorously [3] that right at the lower critical dimension $d = d_c = 2$, the random field destroys the ordered phase, thus the first order transition is rounded by the cluster or domain formation [8]. We are unable to provide such a proof in the case we study here. However heuristically it is easy to see why at $d = d_c$ disorder effects eventually dominate. This is because for a given domain size scale L, there is always some probability that the fluctuation of the number of dopants within a domain is strong enough to overcome the energy penalty of the corresponding domain wall; such probability does *not* decrease with increasing L at $d = d_c$. Therefore domains with arbitrary size will appear at $d = d_c$. We thus expect that for the $1/r$ Coulomb interaction, disorder effects dominate and the first order transitions are rounded by cluster formation in both $d = 2$ and $d = 3$.

(iv) Thus far we have not explicitly discussed the nature of the phases that are involved, and how they may affect the consideration above. If both of the competing phases are insulating and have *no* screening ability, then all of our discussion will just come through with no modification [9]. The situation is more complicated and interesting when metallic phases are involved. Metallic phases have screening abilities; they can maintain charge neutrality of the system at large scales by creating *local* carrier density deviation near a charged impurity to compensate for the its charge; *locally*, the carrier density may deviate from its average value significantly and may even move across the phase boundary. Thus when *both* phases are metallic, the charge imbalance discussed above may be compensated for by screening processes, and our conclusions do not apply beyond the screening length. This is *not* the case however when *one of the two competing phases is insulating*; this is because the insulating phase does not have the screening ability, thus cluster formation cannot be avoided this way when the phase boundary is approached from the insulating side, and the transition from an insulator to a metal is a percolation transition. Even when both phases have screening abilities, our results are still relevant when the screening length ξ is long compared to microscopic length scales (typical for " poor" metals). In this case clusters will form at $d = 3$ up to size $L \sim \xi$ no matter how weak the dopant fluctuation is.

We emphasize that the considerations presented here are rather generic, and the results should be relevant to any first order phase transitions driven by change of doping level in electronic systems. As *possible* examples of the situation discussed here, we now turn our discussion to two electronic systems that are of tremendous current interest, namely the manganites with colossal magneto

resistance and high T_c cuprates. Both compounds have very rich phase diagrams, and in particular, there is a transition from an insulating phase to a metallic phase as dopant concentration is varied (in the latter case the metallic phase is superconducting at low temperature). Recently there has been considerable experimental evidence suggesting that clusters of both phases co-exist and the system becomes inhomogeneous, when the transition is approached [10–17]. It has been suggested that such inhomogeneity is induced by disorder near an otherwise first order transition [18–22]. In the case of the manganites, convincing arguments have been advanced that the cluster formation and the percolative nature of the metal-insulator transition may be the origin of the colossal magnetoresistance [18–20,22], and prediction of possible similar behavior in the cuprates has been made based on the similarity between these two systems [22]. We note that the manganites are 3D systems, which is *above* the lower critical dimension of the RFIM, $d_c = 2$. The cuprates are quasi-2D systems; however its large distance behavior must be determined by the 3D nature of the system. Thus without long-range interactions, the formation of clusters and the resultant inhomogeneity due to disorder near a first order transition is possible but would *not* be guaranteed in either case [23]. The importance of the long-range nature of the Coulomb interaction, as we discussed here, is to *guarantee* cluster formation and inhomogeneity by pushing the lower critical dimension up to $d_c = 3$. Our results are thus consistent with the phenomenology of both the manganites and cuprates, and point to the generality of cluster formation near first order phase transitions in electronic systems, which is of central importance in the theory of Dagotto and co-workers on the colossal magnetoresistance in the manganites, based on phase separation and cluster formation.

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